Dear Dr. Chapman,

we would like to thank you and the 2 anonymous reviewers for their comments which helped us to improve our manuscript. Please find a point-by-point responses to all comments in this document. The line numbers mentioned by you and the reviewers refer to the original version of the manuscript while the line numbers in our responses refer to the revised version of the manuscript.

EDITOR:

Comment 1: This is an easy-to-read paper that synthesizes a large number of experiments on the effects of increasing CO2 on diatom communities. Unfortunately, as the authors state, there is no clear standard for experimental design in such experiments, so it is hard to draw definitive conclusions. This is particularly the case as regards the size of any incubation chamber, where wall effects are known to be important and nutrient depletion may affect results, although large diatoms seem to profit more than smaller ones. It will certainly be of interest to researchers in the field.
REPLY: We thank the Editor for the kind words.

Comment 2: Readers may be somewhat confused by the determination of the "relative degree of realism" (RDR) that the authors use. Some of the writing in this section is a little clumsy, especially lines 196-199, and I could find no reference to this statement in Ferguson et al., 1984, even though it is referenced here.
REPLY: We reformulated many parts of this section to hopefully be clearer in the revised version of the manuscript (e.g. lines 176 – 191).
In line 180 we cite the Ferguson et al. (1984) paper for their finding that confinement of natural bacteria communities in 4 L bottles shifted the community composition towards culturable species. This certainly is an experimental artefact due to confinement so we think this citation is correct.
In line 212 we cite the Ferguson et al. (1984) paper for their finding that differential filtration of the community before confinement altered bacteria growth rates and community composition (they tested 3 μm vs 0.2 μm filtration). They write: "The 3.0 μm filtration increased growth rate and ultimate numbers of culturable cells through the removal of bacterial predators or the release of primary amines from cells damaged during filtration or both". We think that also this citation is correct.
In line 222 we cited the Ferguson et al. (1984) paper for the idea that bottle effects should decrease with increasing V/S. This is only implicit in their paper so that we remove the quotation here.

Comment 3: Also, why does RDR depend on the cube root of the applied mesh size? I can understand why the authors want to reduce the RDR as mesh size decreases, but this definition seems completely arbitrary.
REPLY: The rationale for calculating the cube root of d_{mesh} was that in this case the influence of V/S and d_{mesh} on RDR becomes roughly similar. We added this information to the revised version of the manuscript (lines 249 - 250).

Comment 4: Regarding the figures, Fig. 2 seems unnecessary as all the numbers are stated in the text, and Fig. 4B is not actually discussed; lines 263-264 and the rest of the paragraph actually discuss Fig 4C.

REPLY: We took out Fig. 2 from the revised version of the manuscript. Fig. 4B is indeed not discussed but is nevertheless important for completeness because it shows the fraction of studies that did not report changes in community composition. Fig. 4C is better for the discussion as it more clearly illustrates the specific changes in community composition that have been observed when this was investigated.

REVIEWER #1:

Major points :

Comment 5: Conceptually, I have major issues with the RDR score that indicates higher 'degree of realism'. If looking at the effect of CO2 on diatom physiology, I would rather trust small volume experiments where most conditions are controlled, and with the least amount of other species and grazers and which may show a direct effect on either photosynthesis, growth rates or silicification. The larger the volume, the larger the amount of uncontrolled factors that may impact diatom physiology and community shifts. Selective grazer selection of diatoms (from virus, bacteria, parasites to larger grazers) is probably the largest problem. In a large but confined mesocosm, grazer activity is in my opinion the factor that will control diatom community structure rather than CO2. The larger the volume, the larger are the probabilities of getting different organisms in the control and the various treatments that may confound interpretation.

REPLY: The goal of our study was not to investigate the physiological response of diatoms to OA. Such meta analyses have already been made (Dutkiewicz et al., 2015). Our goal was to summarize how diatoms respond to OA in their natural habitat. More generally, experiments with natural ecological communities (as in our study) do not so much aim for a mechanistic understanding of a certain process (as e.g. in physiological experiments) but rather assess the general sensitivity of more natural communities to environmental drivers. Therefore, (as also noted by the Reviewer below) it is important to have a realistic setup because the net response is composed of a direct physiological response to CO_2 AND by CO_2-induced alterations of interactions with other species. It is therefore, in our opinion, not desirable to exclude important ecosystem components, even if they make interpretations of the results more difficult. While we agree with Reviewer #1 that grazers largely control diatom community structure, the differences in diatom community structure between treatments must still be due to CO_2 because CO_2 is the only factor different between treatments. Therefore, if CO_2 effects on diatoms are indirectly mediated, e.g. by changes in competition with other phytoplankton or via changes in the grazer community, it is likely that these factors are also altered by OA in the real ocean. In our analysis we are interested in the differences between treatments and not primarily in the factors that mostly control diatom community structure as such. Indeed, we extensively discuss in section 4.2 how CO_2 can induce the observed responses in diatom community structure and highlight the potential role of grazers.

Comment 6: The second other confounding parameter would be competition between primary producers. In natural water experiments, the nutrient ratio for instance, together with absolute concentrations may change rapidly and induce indirect changes in community and abundances without any role of CO2. How these confounding factors are deconvoluted in large mesocosms is not clear at all (and in my view impossible).

REPLY: As indicated in the previous reply, a major prerequisite of these kind of experiments is that the initial conditions in all treatments within any given experiment are as identical as possible. Like for grazers, we agree that the nutrient ratio may also play an important role in influencing the diatom community composition as such but this is not the point here. Important for our analysis is that we compare control and high

CO_2 treatments and in this case, CO_2 must be the driving force for the observed difference because all other aspects are the same in between treatments. How exactly CO_2 induces the differences between control and treatment can then differ depending on the physiological CO_2 responsiveness of the diatom community and the ecological settings in the experiment. However, the same applies in nature, where CO_2 increases but other environmental factors (such as nutrient ratios, but also temperature, light etc.) will strongly differ among regions. The point of our study was to account for this natural variability when assessing the direction and magnitude of CO_2 effects on diatom communities – even though it is not possible to deconvolute this (as mentioned by the Reviewer).

Comment 7: I agree that mesocosms are interesting because they take into account trophic relationships and give an insight into net effects of acidification on the total community, but I don't think their RDR scores should be considered more reliable if you're looking at direct impact of CO2 on diatoms only.
REPLY: We are not looking at the direct (i.e. the physiological) impact of CO_2 on diatoms but on the net response within their natural habitat. This includes direct physiological effects as well as indirect effects that are mediated e.g. via competition and trophic interactions. For this purpose, setups yielding a higher RDR score should provide a more reliable picture of the diatom response to OA in the given environment because they arguably (section 2.3) simulate the natural habitat more realistically (although we acknowledge that also larger volumes are associated with artefacts, lines 200 - 207).

Comment 8: What you mention on line 297 « Among these 3, only Pseudo-nitzschia was fairly consistently identified as a "loser" within the investigated natural diatom communities. The relatively weak performance of Pseudo-nitzschia spp. was somewhat surprising because previous monoclonal experiments with this genus often reported a sometimes pronounced positive (Sun et al., 2011; Tatters et al., 2012), or no influence of high CO2 on their growth rate (Sugie and Yoshimura, 2013; Trimborn et al., 2013) but more rarely a negative one (Tatters et al., 2013) » would indeed show that CO2 effects on unique strains show different results than mesocosms experiments.
REPLY: The *Pseudo-Nitzschia* result was taken out from the manuscript (see comment 20). Nevertheless, we want to emphasize that CO_2 effects can play out differently in a monoclonal culture than in a natural habitat. In fact, this is one important point that we want to emphasize with our study (lines 485 – 494). In the natural habitat, CO_2 effects can be induced indirectly through CO_2 effects on competitors and/or grazers. These CO_2 effects on competitors and/or grazers can then lead to a chain reaction of trophic changes at the end of which the tested diatom would show a different response to what we would have expected on a purely physiological response observed in a monoclonal cell culture. We have discussed this in sections 4.1 and 4.2.

Comment 9: I would place the highest reliability of the effect of OA on diatoms on small volume experiments, which have the least amount of trophic levels, with monospecific experiments being the best, and why not competition experiment between 2 diatoms strains.
REPLY: We disagree with the Reviewer's conclusion. We think that the message must not be that smaller scale incubations with no/little ecological context are our best tool to project diatom responses to OA. Stephen Carpenter, who without doubt has the authority to make these statements, has written a seminal paper with the title:

"Microcosm experiments have limited relevance for community and ecosystem ecology" (Carpenter, 1996). Based on insights from 'whole lake experiments' Carpenter nicely illustrates that: "Without context of appropriately scaled field studies, microcosm experiments become irrelevant and diversionary". Furthermore, Carpenter concludes: "Misleading inferences are greatly reduced by scaling research tools to the spatial and temporal extent of ecological processes". Accordingly, we need experiments on different scales, ranging from small-scale monoclonal experiments to large-volume mesocosm studies, and even field studies, to get a reliable picture of how natural systems function and assess their sensitivity to environmental drivers.

Comment 10: This being said, such a literature is much needed and would give valuable insights, but I don't agree at all with the data treatment method presented here, I further think this over- simplification is quite dangerous, and will likely motivate other scientists to run the same kind of "easy" methods to do literature reviews such as this one. This kind of review needs to be done carefully and with a lot of scrutiny. What we need are standardized protocols to look at OA's role on diverse organisms, and push scientists to better conceive their experimental designs, but not to make people think that we can achieve clear conclusions regarding a group as diverse as diatoms from all kinds of mixed experiments, most of which were probably not designed to investigate diatoms specifically.

REPLY: As argued in the previous replies, we think that an over-simplification would be not to account for the different scales of experiments at all. Nevertheless, our analysis includes not only the RDR weighted approach but also the simpler type of analysis where we just counted the number of outcomes and added them to yield a cumulative score. Both analyses ultimately lead to the same conclusions so that the RDR approach does not lead to any major surprises.

To clarify our stand point we added the following section to the revised version of the manuscript: "Finally, we want to point out (and explicitly acknowledge) that the RDR approach to balance the influence of studies on the final outcome of the literature analysis is of course not the one perfect solution and most likely incomplete (see above). However, balancing a literature analysis with the RDR score may still be an improvement relative to the other case where each experiment is treated exactly equal despite huge differences in the experimental setup. Nevertheless, to account for both views (i.e. the RDR is useless vs. the RDR is useful) we will present the outcome of our literature analysis in two different ways throughout the paper: 1) by simply counting the number of outcomes and adding them to yield a cumulative score (N-based approach; left columns in Figs. 3 and 4); 2) by adding the RDR score of the experiments with a certain outcome to yield a cumulative ∑RDR score (RDR-based approach; right columns in Figs. 3 and 4). "

Comment 11: I would welcome this review of papers, but the data needs to be presented more objectively. It is a good idea to regroup experiments by incubation volumes - small volume (1-4 L), middle volume (5-20 L), minicosms, mesocosms- and presence or not of grazers (through mesh size), but not with this weighing method.

REPLY: We have tried to also present results in this incremental manner suggested by the Reviewer but the number of studies is often too low within the same increment to get to any useful conclusions. We also want to point out that grazers cannot be excluded through filtration because grazers are often within the same size class of diatoms (e.g. ciliates).

Minor points:

Comment 12: line 24: what exactly do you lean by positively? higher growth rates, abundance, biomass ?

REPLY: Good point. We meant positively with respect to abundance and biomass. We added this information to the revised version of the manuscript (line 29).

Comment 13: line 53 : since the title is « ocean acidification » I would expect an estimate of the number of marine diatom species here, not total. Sournia et al 91 recognized between 1,400 and 1,800 marine species, more recently the Tara-Oceans metabarcode data revealed up to 4,748 operational taxonomic units (OTU) (Malviya et al 2016), so 30,000 species seem a bit high and probably includes freshwater species ?

REPLY: We thank the Reviewer for pointing this out. The number included freshwater species. We modified this part to be clearer and included the references mentioned by the Reviewer (lines 60 - 66).

Comment 14: line 55 : you may write <2 µm, (e.g. Minutocellus and Minidiscus genera)

REPLY: We googled for the minimum sizes of Minutocellus and Minidiscus and could not find a reference clearly showing they (or some species of these genera) are smaller 2 μm. We found smaller 3 but not smaller 2 μm. We also frequently observed Minidiscus and Arcocellulus in mesocosm studies but neither of them was smaller 2 μm (as we checked with scanning electron microscopy; e.g. Bach et al., 2017). We therefore would prefer to stick to our more conservative estimate.

Comment 15: line 199/203 : I don't understand why you would assume spherical shape for large volume incubators, most are cylinder shaped, with a cone at the base or not depending on the model. I have not yet seen one spherical large volume incubator.

REPLY: The problem of unknown shape does not only apply for mesocosms but also for all the small scale incubations (bottles). Because container shape is not given in most studies (in particular not in the bottle experiments) we assumed the simplest shape (sphere) for all incubations. We justified this in the revised version of the manuscript by stating: "The assumption of spherical shape was necessary because it allowed us to calculate V/S from only knowing V which is usually the only parameter provided with respect to container characteristics. We are aware that this is a simplification because the majority of containers used in experiments will likely have had cylindrical shape. However, the conversion from volume to surface assuming cylindrical shape would have required knowledge of two dimensions (radius and height of the cylinder). Although shape can influence processes within the container (Pan et al., 2015), it is a less important factor to consider in our study because sensitivity calculations assuming reasonable cylinder dimensions showed that the V/S differences due to container shape will be small compared to the V/S differences due to the range of container volumes compared here." (Lines 227 – 236)

Comment 16: line 263 : please correct « ,. »

REPLY: We thank the Reviewer for spotting this mistake and changed accordingly.

Comment 17: line 263 : I believe bibliographical references are not correctly annotated in the text, example : « microscopy except for (Endo et al., 2015) who used molecular tools »

REPLY: We thank the Reviewer for spotting this mistake and changed accordingly. (Also elsewhere).

Comment 18: Table 1 : I find the text too small to read, in particular if printed.
REPLY: The format of this table will be adjusted by the publisher's editing team if the paper is accepted.

REVIEWER #2:

Comment 19: General comment: I have reviewed this manuscript previously and I see there are many changes relative to its earlier version. I really like this idea to summarize the responses of the natural diatom assemblage from Ocean Acidification experiments and this kind of review will help to improve our current understanding and hence will be definitely helpful in modifying future experimental plans.
REPLY: We thank the Reviewer for the kind words.

Comment 20: However, I am only afraid that the number of studies that are considered here (69) are too small with quite large variability in the protocol used for different experiments. This may lead to wrong interpretation of the results. For example, the authors identified Pseudo-Nitzschia as a looser which is highly contradictory to the existing literature on monospecific culture. Moreover, we have conducted some onboard incubation experiments (manuscript under preparation) in the tropical waters and noticed the opposite trend. We found that Nitzschia and Pseudo-Nitzschia are dominating species under increasing CO2 levels. In the paper cited here by the authors (Biswas et al. 2017), the plots for community composition showed that Pseudo-Nitzschia abundance increased and there was no sign of decrease under high CO2 levels. Therefore, I feel that drawing a conclusion based on a limited number of studies could be largely misleading.
REPLY: We understand the Reviewer's concerns and removed the results/discussion on specific taxa from the revised version of the manuscript.

Comment 21: And most importantly, the community shift in relation to increasing/decreasing CO2 levels can be largely dependent on the initial community that is used for the experiment. The paper by Eggers et al (Global change biology, 20(3), 713-723) very clearly demonstrated that the initial community is a key driving force rather than CO2 in the incubations experiments with natural community.
REPLY: Yes, the CO_2 response depends on the tested community enclosed at the beginning of each experiment. We discuss in detail how CO_2 responses can be expressed (through a "direct" physiological response or "indirectly" through altered ecological interactions (see sections 4.1 and 4.2)). We would like to emphasize, however, that all controls and treatments of the investigated experiments initially had enclosed the same communities. Thus, the (significant) differences that occurred in the course of an experiment could only be caused by CO_2. We are interested in this difference and not if another factor is more or less important in determining diatom community composition. (See also our replies to comments 5 and 6).

Comment 22: Further, "different experimental volume" can be a major factor that finally controls the community shift. I am not sure if it would be logical to generalize the responses of the community under very different experimental exposure which would definitely neglect the bottle effect.

REPLY: We agree with the Reviewer which is why we came up with the RDR approach to balance the influence of smaller and larger volume incubations as they are likely associated with a different degree of bottle effects. We point out in the revised manuscript that results from a simple cumulative approach AND the RDR approach are shown in this analysis (lines 279 – 289).

Comment 23: Moreover, the total number of experiments considered here are only 69 including open ocean (28%), coastal (46%), estuarine (16%) and benthic (6%). My question is, can we compare the responses of the diatoms from open ocean and estuarine region, since they have quite different physiology. The former group is never exposed to high CO2 and the later is well acclimatized to a large range of pH variability. I am not sure if this would be really logical to put them in the same scale for a comparison. Pseudo-Nitzschia from open ocean region can be CO2 sensitive, whereas, the same genera from a coastal or estuarine region can be highly resilient. If we do such comparison, then the responses need to be discussed considering their background.
REPLY: We agree with the Reviewer that diatoms may have different sensitivities to OA depending on whether or not they originate from an environment with highly variable CO_2 concentrations. We added an analysis where we separately looked at the bulk diatom response in near shore habitats (coastal + estuarine + benthic) as opposed to oceanic habitats. Our analysis indicates that oceanic diatom communities respond more frequently to high CO_2 than communities from near shore habitats. This is in line with the common notion that OA could have larger effects on open ocean communities (Duarte et al., 2013). We added this analysis to the results (lines 325 – 345, Fig. 4) and discussed them in the revised version of the manuscript (lines 362 – 377).

References for the replies:

Carpenter, S. R.: Microcosm Experiments Have Limited Relevance for Community and Ecosystem Ecology, Ecology, 77(3), 667–680, 1996.

Duarte, C. M., Hendriks, I. E., Moore, T. S., Olsen, Y. S., Steckbauer, A., Ramajo, L., Carstensen, J., Trotter, J. A. and McCulloch, M.: Is Ocean Acidification an Open-Ocean Syndrome? Understanding Anthropogenic Impacts on Seawater pH, Estuaries and Coasts, 36(2), 221–236, doi:10.1007/s12237-013-9594-3, 2013.

Dutkiewicz, S., Morris, J. J., Follows, M. J., Scott, J., Levitan, O., Dyhrman, S. T. and Berman-Frank, I.: Impact of ocean acidification on the structure of future phytoplankton communities, Nat. Clim. Chang., 5(11), 1002–1006, doi:10.1038/nclimate2722, 2015.

Remarks:

All changes in the text are marked in yellow.

CO_2 effects on diatoms: A Synthesis of more than a decade of ocean acidification experiments with natural communities

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Short summary

Diatoms are a group of phytoplankton species responsible for ~20% of primary production on Earth. Ocean acidification (OA) could influence diatoms but the key question is if they become more or less important within marine food webs. Here we synthesize OA experiments with natural communities and found: Diatoms are more likely to be positively than negatively affected by OA and larger species may profit more than smaller ones. This has important implications for ecosystem services diatoms provide.

Abstract

Diatoms account for ~40% of marine primary production and are considered to be key players in the biological carbon pump. Ocean acidification (OA) is expected to affect diatoms primarily by changing the availability of CO_2 as a substrate for photosynthesis

or through altered ecological interactions within the marine food web. Yet, there is little consensus how entire diatom communities will respond to increasing CO_2. To address this question, we synthesized the literature from over a decade of OA-experiments with natural diatom communities to uncover: 1) if and how bulk diatom communities respond to elevated CO_2 with respect to abundance or biomass; 2) if shifts within the diatom communities could be expected and how they are expressed with respect to taxonomic affiliation and size structure. We found that bulk diatom communities responded to high CO_2 in ~60 % of the experiments and in this case more often positively (56 %) than negatively (32 %; 12 % did not report the direction of change). Shifts among different diatom species were observed in 65 % of the experiments. Our synthesis supports the hypothesis that high CO_2 particularly favors larger species as 12 out of 13 experiments which investigated cell size found a shift towards larger species. Unraveling winners and losers with respect to taxonomic affiliation was difficult due to a limited database. We conclude that OA-induced changes in diatom competitiveness and assemblage structure may alter key ecosystem services due to the pivotal role diatoms play in trophic transfer and biogeochemical cycles.

1. Introduction

The global net primary production (NPP) of all terrestrial and marine autotrophs amounts to approximately 105 petagrams (Pg) of carbon per year (Field et al., 1998). Marine diatoms, a taxonomically diverse group of cosmopolitan phytoplankton, were estimated to contribute up to 25 % (26 Pg C $year^{-1}$) to this number, which is more than the annual primary production in any biome on land (Field et al., 1998; Nelson et al., 1995; Tréguer and De La Rocha, 2013). Thus, diatoms are likely the most important single taxonomic group of primary producers on Earth and any change in their prevalence relative to other phytoplankton taxa could profoundly alter marine food web structures and thereby affect

ecosystem services such as fisheries or the sequestration of CO_2 in the deep ocean (Armbrust, 2009; Tréguer et al., 2018).

The most conspicuous feature of diatoms is the formation of a silica shell, which is believed to primarily serve as protection against grazers (Hamm and Smetacek, 2007; Pančić and Kiørboe, 2018). Since the formation of this shell requires dissolved silicate, diatoms are often limited by silicon as a nutrient rather than by nitrogen or phosphate (Brzezinski and Nelson, 1996). However, when dissolved silicate is available, diatoms benefit from their high nutrient uptake and growth rates, allowing them to outcompete other phytoplankton and form intense blooms in many ocean regions (Sarthou et al., 2005).

Diatoms display an enormous species richness, with recent estimates accounting for so far undiscovered diatoms (including freshwater) being in the range of 20,000 – 100,000 species (Guiry, 2012; Mann and Vanormelingen, 2013). Sournia et al. (1991) derive a number between 1400 – 1800 of described marine diatoms based on microscopy while *Tara* Oceans reported ~4700 operational taxonomic units from genetic samples distributed over all major oceans except the North Atlantic and North Pacific (Malviya et al., 2016). Known diatom taxa span a size range of several orders of magnitude (<5 µm up to a few mm) with a wide range of morphologies and life strategies, e.g. single cells and cell chains, pelagic and benthic habitats (Armbrust, 2009; Mann and Vanormelingen, 2013; Sournia et al., 1991). Accordingly, they should not be treated as one functional group, but rather as a variety of subgroups occupying different niches.

It is well recognized that the global importance of diatoms as well as their diversity in morphology and life style is tightly linked to the functioning of pelagic food webs and elemental cycling in the oceans. For example, iron enrichment experiments in the

Southern Ocean found that a shift in diatom community composition from thick- to thin-shelled species ("persistence strategy" vs. "boom-and-bust strategy") can enhance carbon and alter nutrient export via sinking particles (Assmy et al., 2013; Smetacek et al., 2012). This may not only affect element fluxes locally but enhance nutrient retention within the Southern Ocean and reduce productivity in the north which underlines how important diatom community shifts can be on a global scale (Boyd, 2013; Primeau et al., 2013; Sarmiento et al., 2004). Likewise, the cell size of diatoms can play an important role in transferring energy to higher trophic levels, as the dominance of larger species is generally considered to reduce the length of the food chain and lead to higher trophic transfer efficiency (Sommer et al., 2002). Consequently, understanding impacts of global change on diatom community composition is crucial for assessing the sensitivity of biogeochemical cycles and ecosystem services in the world oceans.

It has become evident that the sensitivity of diatoms to increasing pCO_2 is highly variable, likely being related to specific traits such as cell size or the carbon fixation pathway, as well as interactions with other environmental factors such as nutrient stress, temperature or light (Gao et al., 2012; Hoppe et al., 2013; Wu et al., 2014). However, it is still rather unclear how these species-specific differences in CO_2 sensitivities manifest themselves on the level of diatom communities. This knowledge gap motivated us to compile the presently available experimental data in order to reveal common responses of diatom communities to high CO_2 and thereby assess potential scenarios of shifts in diatom community composition under ocean acidification.

2. Literature investigation

2.1. Approach

Our original intention was to conduct a classical meta-analysis, which would have yielded

the benefit of a quantitative measure of diatom responses to OA, expressed as an overall effect size (i.e. combined magnitude) such as the response ratio. However, our literature analysis revealed a large variability in experimental pCO_2 ranges as well as measured response variables, which cannot be directly compared among each other (e.g. microscopic cell counts, pigment concentrations, genetic tools). These limitations impede data aggregation as required for a classical meta-analysis. Furthermore, experimental setups differed widely in terms of other environmental factors such as temperature, light, and nutrient concentrations, all of which are known to modulate potential responses to pCO_2 (Boyd et al., 2018), thereby further complicating data aggregation for meta-analysis. Therefore, we chose an alternative, semi-quantitative approach where diatom responses to increasing CO_2 are grouped in categories (see section 2.2) and also allows to account for differences in experimental setups, e.g. with respect to container volume. While this approach excludes the determination of effect size, it provides an unbiased insight on the direction of change of potential CO_2 effects.

2.2. Data compilation

We explored the response of diatom assemblages to high CO_2 (low pH) by searching the literature for relevant results with Google Scholar (December 15, 2017) using the following search query: diatom OR Bacillariophyceae AND "ocean acidification" OR "high CO_2" or "carbon dioxide" OR "elevated CO_2" OR "elevated carbon dioxide" OR "low pH" OR "decreased pH". The first 200 results were inspected and considered to be relevant when they were published in peer-reviewed journals, contained a description of the relevant methodological details, a statistical analysis or at least a transparent description of variance and uncertainties, and tested CO_2 effects on natural plankton assemblages (artificially composed communities were not considered). We then carefully checked the cited literature in these relevant studies to uncover other studies that were

missed by the initial search. Furthermore, we checked the "Ocean Acidification news stream provided by the Ocean Acidification International Coordination Centre" under the tag "phytoplankton" (https://news-oceanacidification-icc.org/tag/phytoplankton/) for relevant updates since December 2017 (last check on January 16, 2019).

There were two response variables of interest for the literature compilation:

1) The response of the "bulk diatom community" to high CO_2. For this we checked if the abundance of diatoms, the biomass of diatoms, or the relative portion of diatoms within the overall phytoplankton assemblage increased or decreased under high CO_2 relative to the control. We distinguished between "positive", "negative", and "no effect" following the statistical results provided in the individual references. When the CO_2 effect on the bulk community was derived from abundance data we also checked if there are indications for a concomitant shift in the biomass distribution among species. This is relevant because, for example, an increase in bulk abundance could coincide with a decrease in bulk biomass when the species driving the abundances is smaller. We found no indications for conflicting cases but acknowledge that not every reference provided sufficient data on morphological details to fully exclude this scenario. Furthermore, we emphasize that CO_2 can also shift the temporal occurrence of a diatom response (Bach et al., 2017). For example, a diatom bloom could occur earlier in a high CO_2 treatment than in the control but with a similar bloom amplitude (Donahue et al., 2019). In this case we assigned a "positive" response because an earlier bloom occurrence mirrors a higher net growth rate under elevated CO_2.

2) The CO_2-dependent species shifts within the diatom community with respect to taxonomic composition and/or size structure. Unfortunately, cell size of the species was not reported for all experiments. Thus, we distinguished between "no shifts", "shifts

147 between species with unspecified size", as well as "shifts towards larger or smaller
148 species" when this information was provided. Furthermore, we noted the winners and
149 losers within the diatom communities when these were reported (on the genus level).

150 In case the data was taken from factorial multiple stressor experiments (e.g. CO_2 x
151 temperature) we only considered the control treatment at ambient conditions (e.g. at
152 control temperature). Furthermore, we extracted various metadata from each study
153 largely following the literature analysis of Schulz et al. (2017). All bulk diatom responses,
154 community shifts, and metadata is compiled/described in Table 1 and most of it is self-
155 explanatory (e.g. incubation temperature). The coordinates from where the investigated
156 plankton communities originate are given in Table 1 and illustrated in Figure 2. Their
157 habitats were categorized according to water depth, salinity, or life style in the case of
158 benthic communities: "oceanic" = water depth > 200 m (unless the habitat lies within a
159 fjord or fjord-like strait), S > 30; "coastal" = water depth < 200 m, S > 30; "estuarine" =
160 water depth < 200 m, S < 30; "benthic" = benthic communities (diatoms growing on
161 plates) were investigated. We reconstructed the water depth in case it was not provided
162 in the paper using Google Earth Pro (version 7.3.2.5495). The coordinates provided in
163 some of the experiments conducted in land-based facilities were imprecise and marked
164 positions on land. In this case the habitats were set to coastal or estuarine depending on
165 salinity. If salinity was not given we checked the location on Google Earth for potential
166 fresh water sources and also checked the text for more cryptic indications (e.g.
167 "euryhaline" in a lagoon were strong indications for an estuarine habitat). The methods
168 with which responses of the bulk diatom communities to high OA were determined varied
169 greatly among studies and included light microscopy (LM), pigment analyses (PA), flow
170 cytometry (FC), genetic tools (PCR), and biogenic silica (BSi) analyses (Table 1).

171 **2.3. Accounting for different experimental setups to balance the influence of**

individual studies on the outcome of the literature analysis

The most realistic OA experiment would be one where all aspects of the natural habitat are represented correctly. Such setups are possible for benthic communities which can be sampled *in situ* along a natural CO_2 gradient at volcanic CO_2 seeps (Fabricius et al., 2011; Hall-Spencer et al., 2008; Johnson et al., 2011). However, pelagic communities are advected with currents so that it is very difficult to simulate OA in open waters. Thus, OA experiments where pelagic communities are exposed to increasing levels of CO_2 were so far always performed in closed containers even though it is well known that confinement causes experimental artefacts (Calvo-Díaz et al., 2011; Ferguson et al., 1984; Guangao, 1990; Menzel and Case, 1977). The degree by which confinement causes experimental artefacts will differ from study to study depending on factors such as the incubation volume, the length of incubation, or the selective removal of certain size classes from the incubation (Carpenter, 1996; Duarte et al., 1997; Nogueira et al., 2014). In our literature synthesis we had to deal with a large variety of experimental setups and there are very likely differences how well a given setup represents the natural environment. Therefore, we aimed to develop a metric that allows us to estimate "how well the natural system (which we are ultimately interested in) is represented by the experimental setup". This metric – termed the "relative degree of realism (RDR)" – was used to balance the influence of individual studies on the final outcomes of the literature analysis. Most certainly, we do not mean to devalue any studies but think that the highly different scales of experiments, ranging from 0.8 L lab incubations to 75 m^3 *in situ* mesocosms, should not be ignored when evaluating the literature. In the following we will first derive the equation for the RDR and introduce the underlying assumptions. Afterwards we describe aspects that were considered while conceptualizing the RDR.

The incubation volume in the studies considered herein ranged from bottle experiments

197 to *in situ* mesocosm studies with considerably larger incubation volumes. Smaller
198 differences in incubation volumes (e.g. 0.5 vs. 2 L) were shown to have no, or a minor,
199 influence on physiological rates (Fogg and Calvario-Martinez, 1989; Hammes et al.,
200 2010; Nogueira et al., 2014; Robinson and Williams, 2005). However, they can influence
201 food web composition (Calvo-Díaz et al., 2011; Spencer and Warren, 1996), e.g. by
202 unrepresentatively including certain organism groups such as highly motile
203 mesozooplankton. Larger differences of incubation volumes (e.g. 10 vs. 10000 L) are
204 considered to have a major influence on the enclosed communities, with the larger volume
205 being more representative of natural processes (Carpenter, 1996; Duarte et al., 1997;
206 Sarnelle, 1997). Therefore, our first assumption to conceptualize the RDR was that larger
207 incubation volumes represent nature generally better than smaller ones.

208 Plankton communities were pre-filtered in many experiments to exclude larger and often
209 patchily distributed organisms (e.g. copepods). This is a valid procedure to reduce noise
210 and to increase the likelihood to detect CO_2 effects but it also influences the development
211 of plankton communities since the selective removal of certain size classes can modify
212 trophic cascades within the food web (Ferguson et al., 1984; Nogueira et al., 2014). For
213 example, Nogueira et al. (2014) compared plankton successions of pre-filtered (100 µm)
214 and unfiltered communities and found that the removal of larger grazers and diatoms gave
215 room for green algae and picophytoplankton to grow. Such manipulations make the
216 experiment less representative for a natural food web which brought us to the second
217 assumption for the RDR: The smaller the mesh size during the pre-filtration treatment,
218 the less complete and thus the less realistic is the pelagic food web.

219 To parameterize the two abovementioned assumptions we first converted the volume
220 information provided in each experiment into a volume-to-surface ratio (V/S). The
221 underlying thought is that V increases with the third power to the surface area of the

incubator and is indicative for the relation of open space to hard surfaces. Therefore we first converted V into a radius (r) assuming spherical shape:

$$r = \sqrt[3]{\frac{3}{4}\frac{V}{\pi}} \quad (1).$$

The surface (S) of the spherical volume was calculated as:

$$S = 4\pi r^2 \quad (2)$$

The assumption of spherical shape was necessary because it allowed us to calculate V/S from only knowing V which is usually the only parameter provided with respect to container characteristics. We are aware that this is a simplification because the majority of containers used in experiments will likely have had cylindrical shape. However, the conversion from volume to surface assuming cylindrical shape would have required knowledge of two dimensions (radius and height of the cylinder). Although shape can influence processes within the container (Pan et al., 2015), it is a less important factor to consider in our study because sensitivity calculations assuming reasonable cylinder dimensions showed that the V/S differences due to container shape will be small compared to the V/S differences due to the range of container volumes compared here.

The influence of pre-filtration treatments of the investigated plankton community is implemented by multiplying the V/S with the cube root of the applied mesh size (d_{mesh} in µm) so that the RDR is defined as:

$$RDR = \frac{V}{S}\sqrt[3]{d_{mesh}} \quad (3).$$

Thus, as for V/S, the influence of d_{mesh} on RDR does not increase linearly but becomes less influential with increasing d_{mesh}. The rationale for the non-linear increase is that

243 incubations will still have an increasing bias even if they do not have any pre-filtration
244 treatment due to generally increasing organism motility with size. For example, when
245 collecting a plankton community with a Niskin bottle, more motile organisms can escape
246 from the approaching sampler so that the food web composure is still affected even
247 without subsequent pre-filtration. For this reason we also capped the maximum d_{mesh} to
248 10,000 µm when there was no pre-filtration treatment applied since none of the studies
249 included significantly larger organisms. The rationale for calculating the cube root of
250 d_{mesh} was that in this case the influence of V/S and d_{mesh} on RDR becomes roughly similar.
251 Figure 1 illustrates the change of RDR as a function of V and d_{mesh}. High RDRs are
252 calculated for large-scale *in situ* mesocosm studies (~50 – 190) while bottle experiments
253 yield RDRs between ~1 – 12.

254 The key pre-requisite for an experimental parameter to be included in the RDR equation
255 (eq. 3) was that it is reported in all studies. Many parameters that we would have liked to
256 use for the RDR are either insufficiently reported (e.g. the light environment) or not
257 provided quantitatively at all (e.g. turbulence). We therefore had to work with very basic
258 properties related to the experimental setup rather than to the experimental conditions.

259 A particularly critical aspect of the RDR we had to deal with was the duration of the
260 experiments (Time). Time is reliably reported in all studies and therefore principally
261 suitable for the RDR. Our first thoughts were that a realistic community experiment
262 should be long enough to cover relevant ecological processes such as competitive
263 exclusion and therefore also parameterized Time in the first versions of the RDR
264 equation. However, we decided to not account for it in the final version because the
265 factors that define the optimal duration of an experiment are poorly constrained. For
266 example, a 1 day experiment in a 10 L container could indeed miss important CO_2 effects
267 caused by food web interactions. On the other hand, a 30 days experiment in the same

container could reveal such indirect effects but at the same time be associated with profound bottle effects and make the study unrepresentative for simulated natural habitat. Thus, too long and too short are both problematic and the optimum is hard to find. One such attempt to find the optimum Time was made by (Duarte et al., 1997) who analyzed the plankton ecology literature between 1990 – 1995. By correlating the experimental duration with the incubation volume of published experiments they provided an optimal length for any given volume. However, as noted by Duarte et al. (1997), their correlation is based on publication success and therefore rather reflects common practice in plankton ecology experiments and not necessarily a mechanistic understanding of bottle effects. Thus, as there is no solid ground for a parameterization of Time we ultimately decided to not consider it for the RDR.

Finally, we want to point out (and explicitly acknowledge) that the RDR approach to balance the influence of studies on the final outcome of the literature analysis is of course not the one perfect solution and most likely incomplete (see above). However, balancing a literature analysis with the RDR score may still be an improvement relative to the other case where each experiment is treated exactly equal despite huge differences in the experimental setup. Nevertheless, to account for both views (i.e. the RDR is useless vs. the RDR is useful) we will present the outcome of our literature analysis in two different ways throughout the paper: 1) by simply counting the number of outcomes and adding them to yield a cumulative score (N-based approach; left columns in Figs. 3 and 4); 2) by adding the RDR score of the experiments with a certain outcome to yield a cumulative ∑RDR score (RDR-based approach; right columns in Figs. 3 and 4).

3. Results

We found 54 relevant publications on CO_2 experiments with natural diatom assemblages.

Some publications included more than one experiment so that 69 experiments are considered hereafter (Table 1). Most were done with diatom communities from coastal environments (46 %) and oceanic (28%) environments. Estuarine and benthic communities were investigated in 16 % and 6% of the studies, respectively. 4 % of the studies did not provide coordinates where the samples were taken although the region was reported (Table 1; Fig. 2).

Among the 69 experiments, 23 (33 %, ∑RDR = 595) revealed a positive influence of CO_2 on the "bulk diatom community" (see section 2.2), while 13 (19 %, ∑RDR = 266) revealed a negative one. 5 experiments (7 %, ∑RDR = 21) found a CO_2 effect but did not specify whether it is a positive or negative one. 28 experiments (41 %, ∑RDR = 706) found no effect (Fig. 3A).

We also checked if the pCO_2 range tested in the experiments had an influence on whether the bulk diatom community responded to changing carbonate chemistry. This was done because we expected the likelihood to find an OA response to be higher when the pCO_2 difference between treatments and controls is larger. Thus, we calculated the investigated pCO_2 range (highest pCO_2 – lowest pCO_2) for each experiment and categorized the range into "small" (≤300 µatm), "medium (300 – 600 µatm), and "large" (≥600 µatm). Among the 41 experiments that found a CO_2 effect on the bulk diatom community (positive, negative, and unreported direction of change), 4 (10 %, ∑RDR = 106) found it within the low range, 12 (32 %, ∑RDR = 123) in the medium range, and 25 experiments (68 %, ∑RDR = 653) in the high range. Among the 28 experiments that found no CO_2 on the bulk diatom community, 3 (12 %, ∑RDR = 12) tested within the low range, 8 (32 %, ∑RDR = 230) within the medium range, and 17 experiments (68 %, ∑RDR = 465) within the high range. According to this analysis, the likelihood of detecting a CO_2 effect on the

bulk diatom community does not depend on the investigated pCO_2 range.

CO_2-dependent shifts in diatom species composition were investigated with light microscopy except for Endo et al. (2015) who used molecular tools. Species shifts were investigated in a subset of 40 of the 69 experiments (Fig. 3B). Within this subset of 40 studies, 12 (30 %, ∑RDR = 265) found a shift towards larger diatom species under high CO_2, 1 (2.5 %, ∑RDR = 10) found a shift towards smaller diatom species, and 13 (32.5 %, ∑RDR = 67) found no CO_2 effect on diatom community composition. 14 studies (35 %, ∑RDR = 141) reported a CO_2-dependent shift but did not further specify any changes in the size-class distribution (Fig. 3C).

We also tested if the bulk diatom response to OA in coastal, estuarine, and benthic environments was different from the bulk response in oceanic environments. The rationale for this comparison was that carbonate chemistry conditions in oceanic environments may generally be more stable than in the often more productive coastal, estuarine, and benthic environments (Duarte et al., 2013; Hofmann et al., 2011). Therefore, diatoms from oceanic environments may be more sensitive to OA (Duarte et al., 2013). We found 47 experiments with coastal + estuarine + benthic diatom communities. Within this subset, 15 experiments (32 %, ∑RDR = 557) revealed a positive influence of CO_2 on the "bulk diatom community" while 6 (13 %, ∑RDR = 244) revealed a negative one. 4 experiments (9 %, ∑RDR = 19) found a CO_2 effect but did not specify whether it is a positive or negative one. 22 experiments (47 %, ∑RDR = 693) found no effect (Fig. 4A). In contrast, we found 19 experiments with oceanic communities. Within this subset, 5 experiments (26 %, ∑RDR = 17) revealed a positive influence of CO_2 on the "bulk diatom community" while 7 (37 %, ∑RDR = 21) revealed a negative one. 1 experiment (5 %, ∑RDR = 2) found a CO_2 effect but did not specify whether it is a

positive or negative one. 6 experiments (32 %, ∑RDR = 13) found no effect (Fig. 4B). Overall, we found a bulk diatom response to OA (positive, negative, and unreported direction of change) in 53 % of the experiments in coastal + estuarine + benthic environments as opposed to 68 % in oceanic environments. Thus, an OA response of the bulk diatom community was more frequently observed in oceanic environments which was mostly due to the higher frequency of negative OA responses (Fig. 4).

4. Discussion

Numerous physiological studies have shown that diatom growth and metabolic rates can be affected by seawater CO_2 concentrations, and that these responses vary widely among different species (Gao and Campbell, 2014). Such inter-specific differences in pCO_2 sensitivity are an important feature as this could alter the composition of diatom assemblages in a changing ocean. In this regard, it is interesting to note that paleolimnologists have long been using diatom species composition as paleo-proxy to reconstruct lake pH (Battarbee et al., 2010). Hence, there is ample evidence that high CO_2 conditions have the potential to change the diatom species composition.

Indeed, our analysis revealed that CO_2-induced changes in diatom community composition occurred in 27 out of 40 (i.e. 68 %) of community-level experiments which investigated species composition (Fig. 3C). This is certainly a conservative outcome because many studies have only looked at dominant species. In fact, one of the few experiments that investigated the diatom assemblage with higher taxonomical resolution found CO_2 effects also on subdominant species (Sommer et al., 2015) which may have been overlooked in many other experiments.

The comparison of OA effects in different environments revealed that bulk diatom communities responded more frequently to OA in oceanic than in coastal + estuarine +

benthic environments. Especially negative effects of OA were more frequent in oceanic environments (Fig. 4). This result is not particularly surprising since communities found near coasts may be adapted to larger carbonate chemistry variability and therefore be better suited to deal with OA (Duarte et al., 2013). It should be kept in mind, however, that this comparison is based on "only" 19 oceanic experiments in contrast to 47 coastal + estuarine + benthic experiments. Furthermore, our habitat characterization depends on certain criteria (mainly water depth and salinity; see section 2.2) and these may be insufficient for our habitat comparison. For example, plankton communities from near oceanic islands such as the Azores were labelled as "coastal" although they may have been moving within oceanic currents and just happened to be close to shore when they were collected. Accordingly, this type of habitat comparison would be more robust if the community had been characterized based on the prevailing carbonate chemistry they are usually exposed to. Unfortunately, information on the background carbonate chemistry is hardly ever provided.

4.1 CO_2 effects on diatom assemblages originating from (direct) physiological responses to high CO_2

Most studies that found effects of pCO_2 on diatom communities related these changes to CO_2 fertilization of photosynthesis. Concentrations of CO_2 in the surface ocean are relatively low compared to other forms of inorganic carbon, especially bicarbonate ion (HCO_3^-) (Zeebe and Wolf-Gladrow, 2001). However, RubisCO, the primary carboxylating enzyme used in photosynthesis, is restricted to CO_2 for carbon fixation and has a relatively low affinity for CO_2 compared to O_2 (Falkowski and Raven, 2007). Therefore, diatoms (like many other phytoplankton species) operate a carbon concentrating mechanism (CCM) to enhance their CO_2 concentration at the site of fixation relative to external concentrations (e.g. by converting HCO_3^- to CO_2) and thereby

389 establish higher rates of carbon fixation than what would be possible when only
390 depending on diffusive CO_2 uptake (Giordano et al., 2005). It is well known that the
391 proportion of CO_2 uptake vs. HCO_3^- uptake for photosynthesis varies largely among
392 diatoms (Burkhardt et al., 2001; Rost et al., 2003; Trimborn et al., 2008) and is
393 theoretically also a function of cell size (Flynn et al., 2012; Wolf-Gladrow and Riebesell,
394 1997). Accordingly, increasing seawater pCO_2 may increase the proportion of diffusive
395 carbon uptake and/or lower the energy and resource requirements for CCM operation
396 (Raven et al., 2011). From a physiological point of view, these mechanisms could allow
397 for increased rates of photosynthesis and cell division.

398 So how do these theoretical considerations align with (A) the variable and species-
399 specific physiological responses of diatoms to increasing CO_2 (Dutkiewicz et al., 2015),
400 and (B) the results from community-level experiments compiled in this study? Regarding
401 the variability of physiological responses, progress has recently been made by Wu et al.
402 (2014) who experimentally demonstrated a positive relationship between cell volume and
403 the magnitude of the CO_2 fertilization effect on diatom growth rates. Their findings agree
404 well with theoretical considerations, which predict that high CO_2 is particularly beneficial
405 for carbon acquisition by larger species as they are more restricted by diffusion gradients
406 due to lower surface-to-volume ratios than smaller cells (Flynn et al., 2012; Wolf-
407 Gladrow and Riebesell, 1997). The outcome of our literature analysis supports this
408 allometric concept (Fig. 3, Table 2). Twelve out of 13 experiments in which cell size was
409 taken into account found a shift towards larger species. This is reflected in the ∑RDR
410 score of 265 which is ~25 times higher than the opposite result (i.e. CO_2-induced shifts
411 towards smaller diatoms, Fig. 3C). An allometric scaling of CO_2 sensitivity is particularly
412 useful for modelling since cell size is a universal trait which is relatively easy to measure
413 and therefore frequently available (Ward et al., 2012). Accordingly, it may lead to

414 significant improvements of ecological and/or biogeochemical model projections under
415 CO_2 forcing when more than one size class for diatoms is considered.

416 However, although the Wu et al. (2014) allometric approach constitutes a solid starting
417 point to help understanding the variable responses of different diatom species, it probably
418 also still needs some further refinements. For example, central components of CCMs
419 seem to be adapted to diatom cell sizes, thereby potentially alleviating a strict cell size
420 dependency of CO_2 limitation (Shen and Hopkinson, 2015). Furthermore, size
421 dependency alone cannot account for taxon-specific differences in the mode of carbon
422 acquisition (diffusive uptake of CO_2 vs. CCM-supported uptake of HCO_3^-) and how this
423 will affect the competitive ability of species under increasing CO_2. OA will lead to much
424 larger changes in dissolved CO_2 than in HCO_3^-. Thus, species that rely to a larger extent
425 on a resource-intensive CCM may benefit more from increasing pCO_2 on a cellular level,
426 as they could increase the proportion of diffusive CO_2 uptake. However, it is also possible
427 that the same species would be disadvantaged on the community-level, because their
428 niche (i.e. being competitive at lower CO_2 due to an efficient CCM) is diminished under
429 high CO_2 conditions. Which of the scenarios occurs in nature would also depend on how
430 flexible species are in terms of switching carbon acquisition modes, as well as resource
431 allocation. In this regard, it is noteworthy that only few physiological studies on OA
432 effects have taken into account the role of changing nutrient concentrations or even a
433 transition to nutrient limitation. The available experimental evidence suggests that
434 increasing pCO_2 may reduce cellular nutrient requirements for CCM operations and
435 therefore free resources for elevated maximum diatom population densities, particularly
436 when running into nutrient limitation (Taucher et al., 2015). Unfortunately, however, the
437 relevance of this mechanism has so far only been investigated in monoclonal laboratory
438 experiments but not on the community-level.

These considerations illustrate that cell size is an important factor, but is not sufficient to predict physiological or even community-level of diatoms to OA. Moreover, the allometric concept as well as the additional mechanisms described above generally presume positive effects of CO_2-fertilization, thus yielding no first order explanations for observed negative responses of diatoms to changing carbonate chemistry. Obviously, increasing CO_2 concentrations are accompanied by increasing proton (H^+) concentrations under ocean acidification. High H^+ concentrations may reduce key metabolic rates above certain thresholds and outweigh the positive influence of CO_2 fertilization as has been observed in coccolithophores (Bach et al., 2011, 2015).

Another pathway by which ocean acidification may alter diatom communities is the pH effect on silicification and silica dissolution. Low seawater pH should theoretically facilitate silicification as the precipitation of opal occurs in a cellular compartment with low pH conditions (pH ~5) (Martin-Jézéquel et al., 2000; Vrieling et al., 1999). At the same time, a lower pH should reduce chemical dissolution rates of the SiO_2 frustule (Loucaides et al., 2012). While experimental evidence on this topic is still scarce and partly controversial (Hervé et al., 2012; Mejía et al., 2013; Milligan et al., 2004), it is not unlikely that OA-induced changes in the formation and dissolution of biogenic silica may alter the strength of the frustule and therefore the palatability of diatoms to zooplankton grazers (Friedrichs et al., 2013; Hamm et al., 2003; Liu et al., 2016; Wilken et al., 2011). As for the other physiological effects e.g. on carbon fixation, it is likely that OA impacts on silicification will vary among different diatoms species e.g. according to their species-specific intrinsic buffering capacity, thereby leading to further taxonomic shifts within diatom communities.

The response of diatoms to increasing pCO_2 in natural environments will be further modified by multiple other environmental drivers changing simultaneously. Climate

change is expected to elevate ocean temperature, as well as also irradiance and nutrient availability via changes in stratification. Physiological experiments have shown that elevated pCO_2 may have beneficial effects under low and moderate irradiance, but this effect may reverse under high light conditions due to enhanced photoinhibition (Gao 2012). Analogously, warming may have positive or negative effects on photosynthesis and metabolism in general, depending on the thermal optima of the respective species (Boyd et al., 2018). Altogether, these multiple additional drivers will also affect diatom communities, leading to shifts in their taxonomic composition and size structure, which will interact with the impacts of OA.

4.2 Indirect CO_2 effects on diatom assemblages through food web interactions

Diatom community responses can not only originate from a direct CO_2 effect on their physiology but also be caused indirectly through CO_2 responses on other components of the food web. For example, if a grazer of a diatom species is negatively affected by OA then this may benefit the prey and indirectly promote its abundance. Direct OA impacts on zooplankton communities are usually assumed to play a minor role, although there is some experimental evidence that lower pH may have physiological effects at least on some sensitive species or developmental stages (Cripps et al., 2016; Thor and Dupont, 2015; Thor and Oliva, 2015). Nevertheless, much of the currently available empirical evidence indicates that zooplankton communities are affected by OA rather via bottom-up effects, e.g. via changes in primary production or taxonomic composition of the phytoplankton community (Alvarez-Fernandez et al., 2018; Meunier et al., 2017). However, bottom-up effects on zooplankton biomass, size structure, or species composition may in turn trigger feedbacks on diatom communities, thereby leading to a feedback loop that may reinforce until a new steady state is reached. Such considerations illustrate that also second or third order effects need to be considered when assessing OA

effects on the level of ecological communities. Accounting for such indirect effects requires a holistic approach considering all key players in of the food web (something that is beyond the scope of this study). Therefore, interpretations about what the observed responses could mean for entire plankton food webs or even biogeochemical element cycles (section 4.3) should always be regarded with some healthy skepticism as they often neglect the potential for indirect effects.

4.3 Implications of changes in diatom community structure for pelagic food webs and biogeochemical cycles

The taxonomic composition and size structure of phytoplankton communities influences the transfer of energy from primary production to higher trophic levels. In theory, larger diatoms should support a more direct transfer because less trophic intermediates are needed and therefore less respiration occurs until prey items are in an appropriate size range for top predators (Azam et al., 1983; Pomeroy, 1974; Sommer et al., 2002). Such a size shift at the bottom of a food web might eventually lead to higher production in higher trophic levels such as fish. Indeed, recent experimental evidence indicated that fish (including commercially important species) could under certain constellations benefit from high CO_2 due to higher food availability, although it was not tested if this response is somehow linked to the diatom size structure (Goldenberg et al., 2018; Sswat et al., 2018).

Fluxes of elements through the oceans are (like fluxes of energy through food webs) influenced by the composition of diatom communities (Tréguer et al., 2018). This is particularly well recognized in the context of organic carbon export to the deep ocean, for which diatoms are considered to play a pivotal role (Smetacek, 1985). Given that high CO_2 favours large and perhaps more silicified diatoms over smaller ones (section 4.1),

513 we might expect accelerated sinking and thus a positive feedback on the vertical carbon
514 flux. This classical hypothesis is supported by observational evidence from two
515 consecutive years of the North Atlantic spring bloom where, despite similar primary
516 production, particulate organic carbon sequestration into the deep ocean was much higher
517 in the year when the larger diatom species dominated (Boyd and Newton, 1995).
518 However, whether the positive relationship between size and carbon export holds under
519 all circumstances is by no means clear (Tréguer et al., 2018) . It is possible that shifts
520 towards larger sized species coincide with shifts in other traits that feed back negatively
521 on carbon export. For example, when the size shift is associated with decreasing C:Si
522 stoichiometry it may ultimately reduce carbon export (Assmy et al., 2013).

523 The abovementioned examples of trophic transfer and export fluxes illustrate the
524 importance of the factor "diatom community structure" in the context of marine food
525 production and biogeochemical fluxes. They also illustrate that our understanding of the
526 feedbacks induced through changes in diatom communities is highly incomplete. Hence,
527 with our limited understanding we can currently not go further than classifying CO_2-
528 induced changes in diatom communities as "a potential risk" that may cause changes in
529 key ecosystem services.

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1010

1011 **Tables and Figures**

1012

1013 **Table 1**. Response of diatom communities to high CO_2. 69 experiments from 54
1014 publications were considered here. Location refers to the place where diatom
1015 communities were collected. The RDR is dimensionless (see methods). T is the average
1016 incubation temperature in °C. DoE are days of experiment with the number of samplings
1017 given as the second number. Pre-filt. gives the mesh size in case the collected plankton
1018 community was pre-filtered before incubation. Setup refers to the incubation style:
1019 undiluted volumes (batch), repeatedly diluted volumes (s.-cont.), flow-through setups

(fl.-thr.; only benthos), chemostats (chem.; only pelagic), CO_2 vent sites (seep; only benthos). Incubations can either be performed on deck (e.g. shipboards), *in situ* (e.g. *in situ* mesocosms) or under laboratory conditions. V refers to the incubation volume. Nutrient ammendments were made in some but not all studies. The element indicates which nutrients were added. Asterisks indicate the presense of residual nutrients at the beginning of the study. Manipulations were done with: CO_2 saturated seawater (SWsat), acid additions (Acid), combined additions of acid and base (Comb.), CO_2 gas additions (CO_2), Aeration at target pCO_2 (Aer.), Passing CO_2 gas through a diffusive silicone tubing (Diff.). Meth. indicates the applied methodology to investigate diatom communities: light microscopy (LM), pigment analyses (PA), flow cytometry (FC), genetic tools (PCR), biogenic silica (BSi). The pCO_2 range of the experiment with the number of treatments given in brackets. The response of the bulk diatom community to CO_2: no effect (~), positive (p), negative (n), not reported (N/A). The pCO_2 response indicates approximately in between which treatments a CO_2 response was observed. Please note that this is based on visual inspection of the datasets and therefore involves subjectivity. Please also note that the range equals the treatment values in case only two treatments were set up. CO_2 induced shifts between diatom species can be: shift to larger species (large), shift to smaller species (small), unspecified shift (shift), no species shift detected (~), not reported (N/A). Winners or losers of the diatom community comprise: *Chaetoceros* (Chae), large *Chaetoceros* (Chae I), medium *Chaetoceros* (Chae II), small *Chaetoceros* (Chae III), *Neosyndra* (Neos), *Rhabdonema* (Rhab), *Eucampia* (Euca), *Cerataulina* (Cera), *Thalassiosira* (Thals), *Proboscia* (Prob), *Pseudo-nitzschia* (Ps-n), *Thalassionema* (Thalns), *Cylindrotheca* (Cyli), *Guinardia* (Guin), *Synedropsis* (Syned), *Dactyliosolen* (Dact), *Toxarium* (Toxa), *Leptocylindrus* (Lept), *Grammatophora* (Gram), *Bacillaria* (Baci), *Navicula* (Navi).

Reference	lat	long	RDR	S	T (°C)	Habitat	DoE/ # of sampl.	Pre-filt. (µm)	Setup	Incub.	V (L)	Nutr.	Manip.	Meth.	pCO_2 range (µatm)	CO_2 effect	pCO_2 response (µatm)	Intra-taxon effect	Winners	Losers
(Bach et al., 2017)	58.264	11.479	76.2	29	7	est.	113/57	1000	batch	in situ	50000	*none	SWsat	PA, LM	(2) 380, 760	p	380 - 760	large	Cosc	
(Bach et al., 2019)	27.990	-15.369	59.6	37	18.5	coastal	32/21	3000	batch	in situ	8000	N,P,Si	SWsat	LM, BSi	(7) 380 - 1120	p	380 - 1120	large	Chae, Guin, Lept	Nitz
(Biswas et al., 2011)	16.750	81.100	2.1	25	29.5	est.	5/2	200	batch	Deck	5.6	*none/N, P	Comb.	PA	(4) 230 - 1860	n	650 - 1400	N/A		
(Biswas et al., 2017)	17.000	83.000	1.5	?	?	coastal	2/1	200	batch	Deck	2	*N,P,Si,Fe,(Zn)	Comb.	LM	(2) 230, 2200	p	230 - 2200	shift	Skel	Thals
(Davidson et al., 2016)	-68.583	77.967	10.5	34	0.1	coastal	8/5	200	batch	Lab	650	*Fe	SWsat	LM	(6) 80 - 2420	n	1280 - 1850	small	Frag	Chae
(Domingues et al., 2017)	37.017	-8.500	7.4	?	23.5	est.	1/1	no	batch	Deck	4.5	N,P,Si,NH4	Comb.	LM, PA	(2) 420, 710	~		~		
(Donahue et al., 2019)	-45.800	171.130	2.6	34	11	oceanic	14/5	200	batch	Lab	10	*Fe	Diff.	LM, FC	(2) 350, 620	~		N/A		
(Donahue et al., 2019)	-45.830	171.540	2.6	34	11	oceanic	21/4	200	batch	Lab	10	*Fe	Diff.	LM, FC	(2) 350, 630	p	350 - 630	N/A		
(Eggers et al., 2014)	38.633	-27.067	1.9	36	15	coastal	9-10/3	200	batch	Deck	4	N,P,Si	Comb.	LM	(2) 380, 910	p	380 - 910	large	Chae III	Thals
(Eggers et al., 2014)	38.650	-27.250	1.9	36	15	coastal	9-10/4	200	batch	Deck	4	N,P,Si	Comb.	LM	(2) 380, 910	p	380 - 910	large	Thals, Chae II	Chae I
(Eggers et al., 2014)	38.617	-27.250	1.9	36	15	oceanic	9-10/5	200	batch	Deck	4	N,P,Si	Comb.	LM	(2) 380, 910	~		N/A		
(Endo et al., 2013)	46.000	160.000	2.8	33	14	oceanic	14/3	197	batch	Deck	12	*none	Aer.	PA	(4) 230 - 1120	~		N/A		
(Endo et al., 2015)	53.083	-177.000	2.8	?	8.2	oceanic	5/3	197	batch	Deck	12	*none	Aer.	PA, PCR	(2) 360, 600	n	360 - 600	~		
(Endo et al., 2016)	41.500	144.000	2.8	?	5.4	oceanic	3/3	197	batch	Deck	12	*Fe	Aer.	PA, PCR	(4) 180 - 1000	n	350 - 1000	shift		
(Feng et al., 2009)	57.580	-15.320	1.7	35	12	oceanic	14/1-2	200	s.-cont.	Deck	2.7	N,P	Aer.	LM, PA	(2) 390, 690	p	390 - 690	large	Ps-n	Cyli
(Feng et al., 2010)	-74.230	-179.230	1.7	34	0	oceanic	18/1-14	200	s.-cont.	Deck	2.7	none	Aer.	LM, PA	(2) 380, 750	~		large	Chae	Cyli
(Gazeau et al., 2017)	43.697	7.312	125.8	38	14	coastal	18/14	5000	batch	in situ	45000	none	SWsat	PA	(6) 350 - 1250	p	600 - 1000	N/A		
(Gazeau et al., 2017)	42.580	8.726	125.8	38	23	coastal	27/18	5000	batch	in situ	45000	none	SWsat	PA	(6) 420 - 1250	~		N/A		
(Grear et al., 2017)	41.575	-71.405	9.3	?	9	est.	6/7	no	chem.	Deck	9.1	?none	Comb.	LM	(3) 220 - 720	~		~		
(Hama et al., 2016)	34.665	138.940	7.1	?	?	coastal	29/11	100	batch	Deck	400	N,P,Si	Aer.	PA	(3) 400 - 1200	~		N/A		
(Hare et al., 2007)	56.515	-164.730	6.0	?	10.4	coastal	9-10/5	no	s.-cont.	Deck	2.5	Fe,N,P, Si	Aer.	LM, PA	(2) 370, 750	n	370 - 750	shift		Cyli
(Hare et al., 2007)	55.022	-179.030	6.0	?	10.4	oceanic	9-10/3	no	s.-cont.	Deck	2.5	Fe	Aer.	LM, PA	(2) 370, 750	n	370 - 750	N/A		
(Hopkins et al., 2010)	60.300	5.200	99.1	?	10	coastal	21/9	no	batch	in situ	11000	N, P	Aer.	LM	(2) 300, 600	n	300 - 600	N/A		
(Hoppe et al., 2013)	-66.833	0.000	1.9	34	3	oceanic	27-30/1	200	s.-cont.	Lab	4	*none	Aer.	LM	(3) 200 - 810	N/A	400 - 810	shift	Syned	Ps-n
(Hoppe et al., 2017b)	71.406	-68.601	1.9	33	9.5	oceanic	8/3	100	s.-cont.	Deck	8	N,P,Si	Aer.	PA, LM	(2) 320, 990	~		~		

(Hoppe et al., 2017a)	63.964	-60.125	1.9	32	7.9	oceanic	13-14/3	100	s.-cont.	Deck	8	N,P,Si	Aer.	LM	(2) 300, 960	n	300 - 960	shift	Frag	Ps-n
(Hussherr et al., 2017)	71.406	-70.188	2.6	33	4.3	oceanic	9/3-9	200	batch	Deck	10	*none	Comb.	LM, PA	(6) 510 - 3300	n	1040 - 1620	~		
(James et al., 2014)	-45.639	170.671		?	11.6	benthic	42/2		fl.-thr.	Lab	0	none	Comb.	pic	(2) 400, 1250	~		N/A		
(Johnson et al., 2011)	38.417	14.950		38	23.5	benthic	21/1		seep	in situ	0	none	NA	PA, LM	(3) 420 - 1600	p	420 - 590	large	Toxa, Gram, Baci, Navi, Cocc	Cycl, Neos, Rhab, Nitz
(Kim et al., 2006)	34.600	128.500	4.3	?	14	coastal	14/?	60	batch	in situ	150	N,P	Aer.	LM	(3) 250 - 750	N/A	400 - 750	shift	Skel	Nitz
(Kim et al., 2010)	34.600	128.500	52.1	?	12	coastal	20/22	no	batch	in situ	1600	N,P,Si	SWsat/ Aer.	LM	(2) 400, 900	~		shift	Skel	Euca
(Mallozzi et al., 2019)	29.241	-90.935	2.4	12	21	est.	112/9	80	s.-cont.	Lab	20	*none	Aer.	PA, LM	(2) 400, 1000	~		shift	Cyli	
(Mallozzi et al., 2019)	29.272	-89.963	2.4	17	21	est.	112/9	80	s.-cont.	Lab	20	*none	Aer.	PA, LM	(2) 400, 1000	~		shift	Cyli	
(Maugendre et al., 2015)	43.667	-7.300	1.9	?	15	oceanic	12/4	200	batch	Deck	4	none	SWsat	PA	(2) 360, 630	~		N/A		
(Nielsen et al., 2010)	56.057	12.648	1.6	19	10.7	est.	14/4	175	s.-cont.	Lab	2.5	*none	Acid	LM, PA	(3) 500 - 1500	~		~		
(Nielsen et al., 2012)	-42.887	147.339	1.8	31	16	coastal	14/4	250	s.-cont.	Lab	2.5	*none	Acid	LM, PA	(3) 300 - 1200	~		~		
(Park et al., 2014)	34.600	128.500	59.6	?	17	coastal	19/17	no	batch	in situ	2400	N,P,Si	SWsat/ Aer.	LM, PA	(6) 160 - 830	p	160 - 830	N/A	Cera	
(Paul et al., 2015)	59.858	23.258	112.7	6	11	est.	46/22	3000	batch	in situ	54000	none	SWsat	PA	(6) 370 - 1230	p	820 - 1000	N/A		
(Reul et al., 2014)	36.540	-4.600	3.3	?	21	coastal	7/6	200	batch	Deck	20	control/N,P	Aer.	LM, PA	(2) 500, 1000	p	500 - 1000	large		
(Roleda et al., 2015)	-45.639	170.671		34	10.8	benthic	112/?		fl.-thr.	Lab	0.65	none	Comb.	PA	(2) 430, 1170	~		N/A		
(Rossoll et al., 2013)	54.329	10.149	8.1	18	18	est.	28/7	200	batch	Lab	300	N,P,Si	Aer.	LM	(5) 390 - 4000	~		N/A		
(Sala et al., 2015)	41.667	2.800	26.1	38	14	coastal	9/2	no	batch	Lab	200	none	CO2	LM	(2) 400, 800	~		N/A		
(Sala et al., 2015)	41.667	2.800	26.1	38	22	coastal	9/2	no	batch	Lab	200	none	CO2	LM	(2) 400, 800	~		N/A		
(Schulz et al., 2008)	60.267	5.217	133.7	31	10.5	coastal	25/18-23	no	batch	in situ	27000	N,P	Aer.	PA	(3) 350 - 1050	~		N/A		
(Schulz et al., 2013)	78.937	11.893	106.1	34	3	coastal	30/26 - 30	3000	batch	in situ	45000	N,P,Si	SWsat	LM, PA	(8) 185 - 1420	~		N/A		
(Schulz et al., 2017)	60.265	5.205	125.8	32	9	coastal	38/35	3000	batch	in situ	75000	*N, P	SWsat	LM, PA	(8) 310 - 3050	n	1165 - 1425	N/A		
(Segovia et al., 2017)	60.390	5.320	99.1	?	11	coastal	22/9	no	batch	in situ	11000	control	SWsat /Aer.	FC	(2) 300, 800	~		N/A		
(Sett et al., 2018)	54.329	10.149	13.5	20	5	est.	44/26	200	batch	Lab	1400	*none	SWsat	LM, FC	(2) 540, 1020	~		~		
(Shaik et al., 2017)	15.453	43.801	5.6	35	29	coastal	2/1	no	batch	Deck	2	N,P,Si,Fe	CO2	LM	(2) 330, 1000	p	330 - 1000	~		
(Shaik et al., 2017)	15.453	43.801	5.6	36	29	coastal	9/1	no	s.-cont.	Deck	2	N,P,Si,Fe	CO2	LM	(2) 400, 1000	p	400 - 1000	~		
(Shaik et al., 2017)	15.453	43.801	5.6	35	29	coastal	2/1	no	batch	Deck	2	N,P,Si,Fe	CO2	LM	(2) 240, 780	p	240 - 780	~		

(Sommer et al., 2015)	54.329	10.149	49.8	20	9,15	est.	24/11	no	batch	Lab	1400	*none	SWsat	LM	(2) 440, 1040	~		shift		Prob, Thaln, Guin, Ps-n, Chae
(Tatters et al., 2013)	-45.752	170.810	0.8	35	14	coastal	14/2	80	s.-cont.	Lab	0.8	N,P,Si,Fe	Aer.	LM	(3) 230 - 570	N/A	400 - 570	shift	Cosc, Ps-n	Navi, Chae
(Tatters et al., 2018)	33.750	-118.215	12.1	?	19	coastal	10/1	no	chem.	Deck	20	N/urea,P, Si	Aer.	LM	380, 800	N/A		shift		
(Taucher et al., 2018)	27.928	-15.365	97.6	37	24-22	coastal	60/35	3000	batch	in situ	35000	N,P,Si	SWsat	LM, PA	(8) 350 - 1030	p	890 - 1030	large	Guin	Lept
(Thoisen et al., 2015)	69.217	53.367	1.4	33	3	coastal	8-17/6-9	250	s.-cont.	Lab	1.2	*none	SWsat	LM	(4) 440 - 3500	n	440 - 900	shift	Navi I	Navi II
(Tortell et al., 2002)	-6.600	-81.017	7.1	?	?	oceanic	11/4	no	s.-cont.	Deck	4	*none	Aer.	PA, LM	(2) 150, 750	p	150 - 440	~		
(Tortell et al., 2008)	NA	NA	7.1	?	0	N/A	10-18/?	no	s.-cont.	Lab	4	*Fe	Aer.	LM, PA	(3) 100 - 800	p	100 - 400	large	Chae	Ps-n
(Tortell et al., 2008)	NA	NA	7.1	?	0	N/A	10-18/?	no	s.-cont.	Deck	4	*Fe	Aer.	LM, PA	(3) 100 - 800	p	100 - 400	large	Chae	Ps-n
(Tortell et al., 2008)	NA	NA	7.1	?	0	N/A	10-18/?	no	s.-cont.	Deck	4	*Fe	Aer.	LM, PA	(3) 100 - 800	p	100 - 400	large	Chae	Ps-n
(Trimborn et al., 2017)	-53.013	10.025	1.9	34	3	oceanic	30/4	200	s.-cont.	Lab	4	none	Aer.	LM	420, 910	n	420 - 910	shift		Ps-n
(Witt et al., 2011)	-23.450	151.917		?	24-25	benthic	11/4		fl.-thr.	Deck	10	none	SWsat	LM	(4) 310 - 1140	p	560 - 1140	N/A		
(Wolf et al., 2018)	78.917	11.933	1.9	?	3	coastal	10 - 13/1	200	s.-cont.	Lab	4	none	Aer.	LM	(2) 400, 1000	N/A	400 - 1000	~		
(Yoshimura et al., 2010)	49.500	148.250	2.7	33	13.5	oceanic	14/5	243	batch	Deck	9		Aer.	PA	(4) 150 - 590	n	150 - 280	N/A		
(Yoshimura et al., 2013)	53.390	-177.010	2.8	?	8.4	oceanic	14/3	197	batch	Deck	12	*none	Aer.	PA, LM	4 (300 - 1190)	p	960 - 1190	N/A		
(Yoshimura et al., 2013)	49.020	174.020	2.8	?	9.2	oceanic	14/3	197	batch	Deck	12	*none	Aer.	PA, LM	(4) 230 - 1110	p	880 - 1110	N/A		
(Young et al., 2015)	-44.779	-64.073	7.1	?	-1	coastal	21/21	no	s.-cont.	Deck	4	*none	Aer.	PA	(3) 100 - 800	~		N/A		
(Young et al., 2015)	-44.780	-64.073	7.1	?	-0.5	coastal	16/16	no	s.-cont.	Deck	4	*none	Aer.	PA, LM	(3) 100 - 800	~		N/A		
(Young et al., 2015)	-44.780	-64.073	7.1	?	1.5	coastal	20/20	no	s.-cont.	Deck	4	*none	Aer.	PA	(3) 100 - 800	~		N/A		

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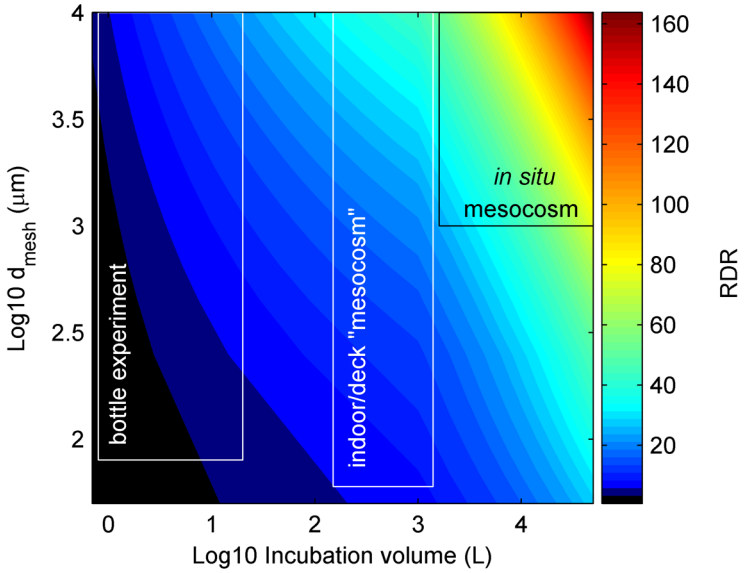
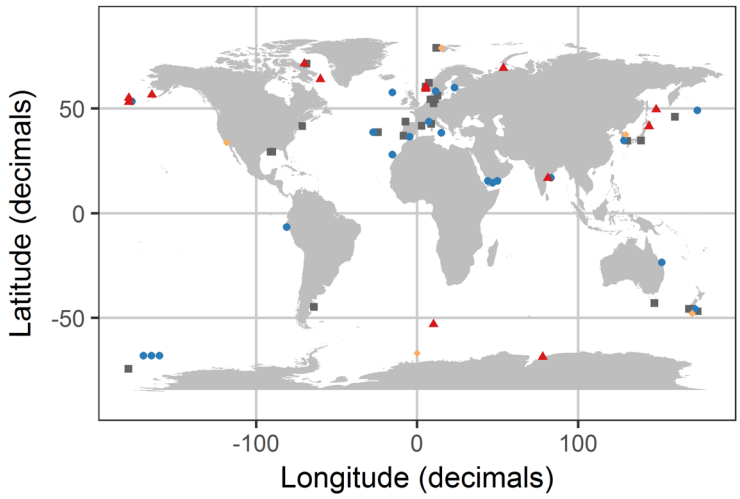


1047

1048 **Figure 1**. RDR as a function of incubation volume and size of the mesh that was used
1049 while filling the incubation volumes (d_{mesh}). The black and white boxes illustrate
1050 approximate ranges of the three main types of containers used in experiments. Please note
1051 that the general definition for mesocosms are volumes >1000 L (Guangao, 1990) but
1052 since most authors also use this term for open batch incubations with volumes between
1053 150 – 1000 L we also stick to this term for the intermediate class.



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1055 **Figure 2.** Distribution of experiments with associated OA response of the bulk diatom

communities as listed in Table 1. Blue circles = positive effect; red triangles = negative response; grey squares = no response; orange diamonds = response not reported. Locations were slightly modified in case of geospatial overlap to ensure visibility. Please note that the three blue points in the Ross Sea at about -68, -165 are approximate locations because the reference did not provide coordinates.

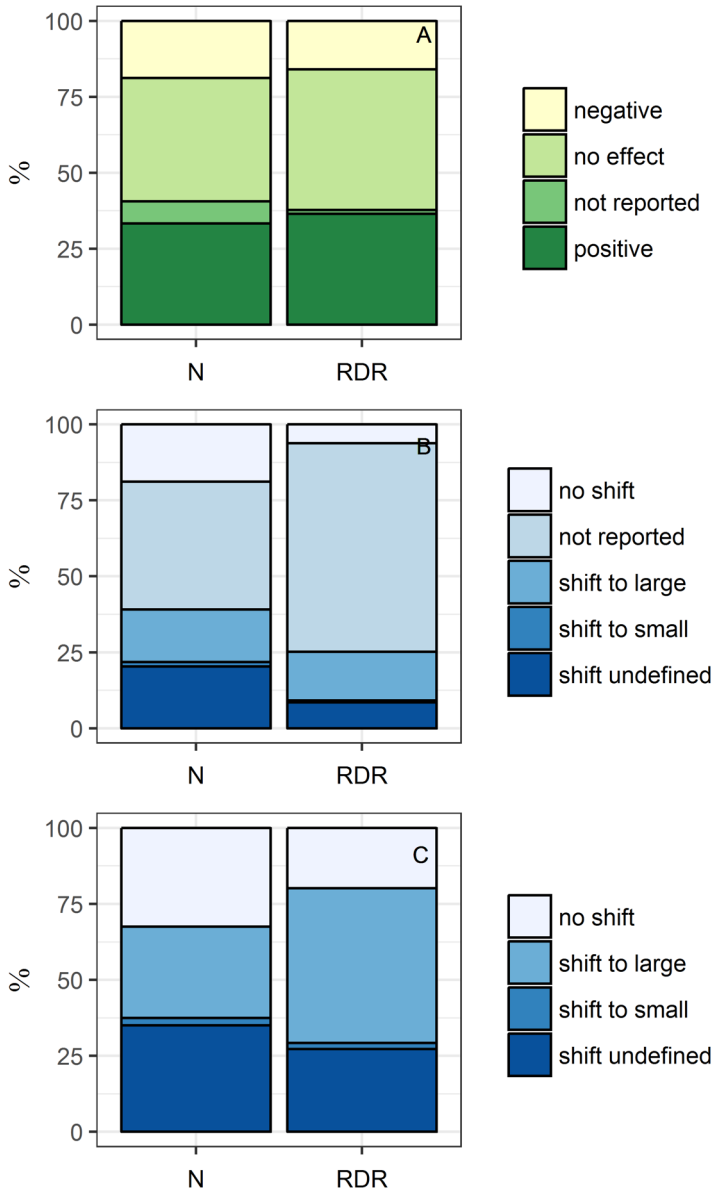

Figure 3. Summary of the literature analysis. (A) Response of the bulk diatom community to ocean acidification. (B) Shifts among different diatom species due to ocean acidification. 'Shift to large' and 'shift to small' indicate that the diatom community

shifted towards the dominance of larger or smaller species, respectively. (C) Same data as in B but excluding studies where species shifts within the diatom community were not reported. This reduced the dataset from 69 to 40 studies. The left column is based on the number of studies. For example, the bulk diatom community was positively affected by OA in 29 out of 69 studies which is 33 %. The right column is based on the RDR values. For example, the ∑RDR value of all studies where the diatom community was positively affected by OA was 605 which is 36 % of the total ∑RDR.

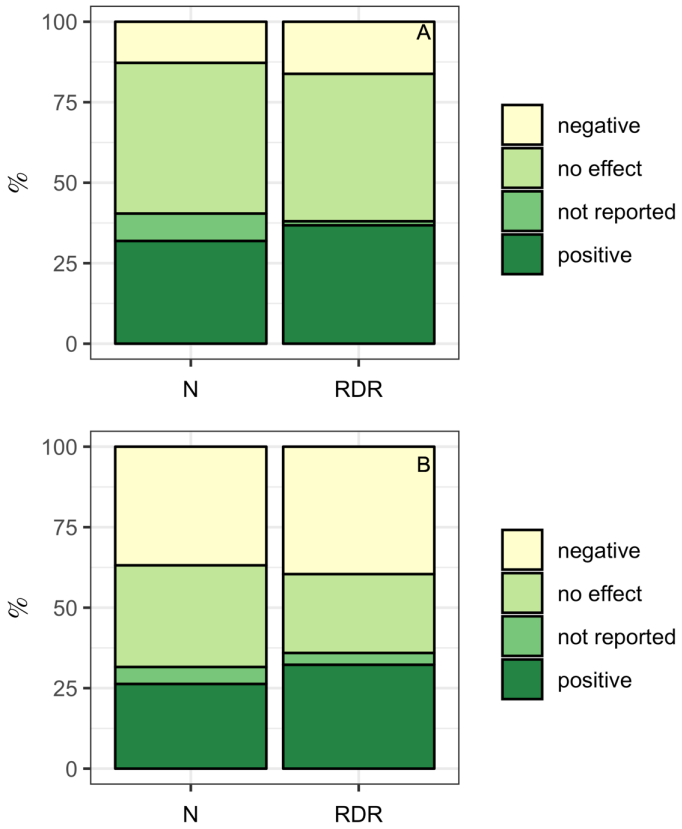

Figure 4. Comparison of the diatom bulk response to OA in different environments. (A) coastal + estuarine + benthic environments with 47 experiments. (B) Oceanic environments with 19 experiments. The left column is based on the number of studies. For example, the bulk diatom community was positively affected by OA in 5 out of 19

1078 studies in oceanic environments which is 26 %. The right column is based on the RDR

1079 values. For example, the ∑RDR value of all studies where the oceanic diatom community

1080 was positively affected by OA was 17 which is 32 % of the total ∑RDR.

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